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Demonstration of laser induced magnetization reversal in GdFeCo nanostructures

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Magnetization switching by a single femtosecond laser heat pulse is demonstrated for out-of-plane domains with sizes down to 200 nm in GdFeCo nanostructures. A complex magnetic domain configuration was revealed with a photoemission electron microscope employing x-ray magnetic circular dichroism at the Fe L₃ edge and consisted of in-plane magnetized rims and out-of-plane domains, which results from the structuring process. No influence of this complex domain pattern on the switching efficiency of the structures was detected, constituting an important step towards the application of laser induced magnetization switching in storage devices. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4733965>]

In order to further increase the recording density of magnetic hard drives, alternative solutions to the magnetic recording trilemma arising from the competing requirements between readability, writeability, and stability have to be found.¹ One promising solution is heat assisted magnetic recording (HAMR) where a laser pulse is used to heat up briefly the magnetic bit to be recorded to lower its coercivity to a value compatible with the magnetic field produced by the recording head.² Recent numerical atomistic scale modeling simulations of the spin dynamics in a Heisenberg GdFe ferrimagnet demonstrated that the rapid transfer of energy into the spin system leads to switching of the magnetization within a few picoseconds without the need of applying any magnetic field. The prediction was confirmed experimentally in GdFeCo ferrimagnetic alloys in which the ultrafast heating was realized with the help of a linearly polarized femtosecond laser pulse.³ This switching via a *heat pulse only* occurs in a ferrimagnet consisting of 3d and 4f metals when the femtosecond laser pulse heats up the free electrons and thus excites the magnet on the time scale of the exchange interaction. This brings the system into a transient strongly non-equilibrium state due to the different demagnetizing time scales of the Gd and Fe sublattices,⁴ which relax deterministically into the reversed state.⁵ Furthermore, using circularly polarized laser pulses, it is possible to take advantage of the magnetic circular dichroic effect to record a magnetic domain in which the final magnetization direction is given solely by the helicity of the laser pulse.⁶ These discoveries suggest that a possibly simpler solution to the magnetic recording trilemma can be proposed, involving only a laser heat pulse instead of the currently used magnetic recording head and displaying tremendous recording speed. It is left to be verified, however, how such switching works in nanostructures, where the initial magnetization states may be strongly modified by magnetostatic fields or by the manufacturing

process itself, and where the relaxation from the transient strongly non-equilibrium state could be altered.

In this letter, we demonstrate the magnetization reversal in GdFeCo nanostructures for domains as small as 200 nm after applying a single, linearly polarized femtosecond laser pulse and without applying any magnetic field, i.e., by a *heat pulse only*. The three components of the magnetization vector in the structures were determined with a photoemission electron microscope (PEEM) by performing an azimuthal dependent measurement of the x-ray magnetic circular dichroism (XMCD) at the Fe L₃ edge. This azimuthal study showed that the rims of the structures display an in-plane magnetic anisotropy, which seems to be the result of the thinning of the GdFeCo layer at the edge resulting from the fabrication process. Moreover, the in-plane rims are found to play no significant role in the reversal of the central out-of-plane domain.

The sample consisted of a thin film of composition AlTi(10 nm)/Si₃N₄(5 nm)/Gd_{24.5}Fe_{66.1}Co_{9.4}(20 nm)/Si₃N₄(3 nm) grown by magnetron sputtering on a silicon substrate. The magnetization compensation temperature was determined to be 370 K with a room temperature coercive field of 28 kA/m with a square out-of-plane hysteresis loop. The structuring of this sample in squares and discs with sizes ranging from $2 \times 2 \mu\text{m}^2$ down to $100 \times 100 \text{ nm}^2$ has been realized via electron beam lithography in combination with a lift-off process, resulting in isolated magnetic structures.⁷ Images of the magnetic domain states in these structures are then obtained using the Elmitec PEEM at the surface/interface: microscopy (SIM) beamline⁸ at the Swiss Light Source. A magnetic image is obtained using the XMCD effect at the Fe L₃ edge, the contrast of which is a quantitative measure of the projection of the element specific magnetic moment on the x-ray polarization vector. For excitation, single laser pulses were selected using a pulse picker in combination with an optical shutter from an XL-500 oscillator from Femtolasers, producing 50 fs laser pulses with 500 nJ per pulse at a 5.2 MHz

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repetition rate. The laser was focused in the PEEM to a spot size characterized by a full width at half maximum (FWHM) of $30 \times 105 \mu\text{m}^2$.

A sequence of XMCD images, each recorded after excitation by a single linearly polarized laser pulse, is shown in Figs. 1(b)–1(e) and illustrates the efficiency of the laser induced magnetization switching in these various structures whose sizes and shapes are shown in the sample layout in Fig. 1(a). For the 2 and $1 \mu\text{m}$ wide structures, the reversal of the magnetization is easily identified as a change from black to white or vice versa with an efficiency approaching 100%. In this specific example, out of the 9 largest structures visible and out of the 3 laser pulses, only one case (circled in Fig. 1(d)) does not result in a complete switching, giving a total switching efficiency of 96%. The behaviour of the smaller structures that are 500, 400, and 300 nm wide is however not as evident as for the larger ones. Here, most of them seem to have stable multi-domain states with both black and white contrast in the same structure, in contrast to the larger structures which look mono-domain. These small structures show an apparently random switching behavior. As will be

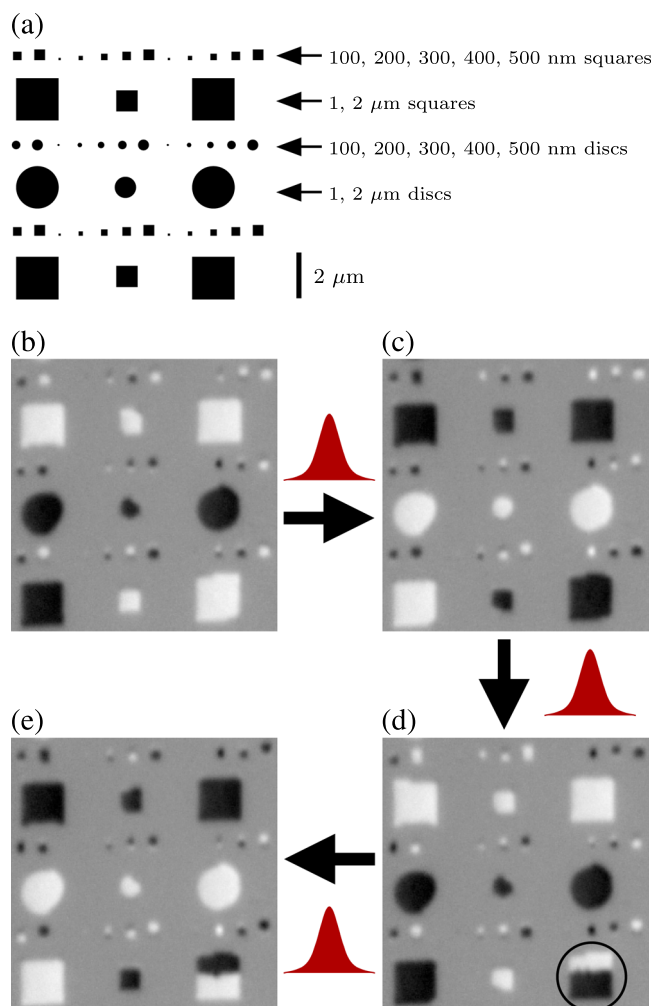


FIG. 1. (a) Sketch of the region of interest showing the various structures sizes and shapes. (b)–(e) Sequence of successive XMCD images from the same region. The arrows indicate the order in which the images were recorded, while a single linearly polarized laser pulse was applied to the sample in between each of them. The circle indicates the case for which the switching is not complete.

shown later, this is due to the structures having in-plane as well as out-of-plane domains which cannot be distinguished in these images and which behave differently. In order to better understand their magnetic configurations and their laser induced switching behavior, an azimuthal dependent study was thus carried out, revealing the spatially resolved magnetization vector components. For this, three different XMCD images of the same region were recorded with three different incoming x-ray azimuthal angles by rotating the sample around the surface normal by 0° , 90° , and 180° . Due to the 16° grazing incidence of the x-rays on the sample, it is then possible to derive the in-plane M_{in} and out-of-plane M_{out} magnetization components.^{9–11}

The derived M_{in} and M_{out} magnetization components from the azimuthal XMCD study of the initial state are shown in Figs. 2(b) and 2(c), respectively. In these images, the color gives the component direction while the brightness gives its magnitude according to the color wheel for the in-plane component and the color double-headed arrow for the out-of-plane component. The azimuthal study was then repeated on the same structures after excitation by a single linearly polarized laser pulse and the result is shown in Figs. 2(d) and 2(e). Comparing the out-of-plane magnetization before and after the laser pulse in Figs. 2(c) and 2(e) clearly shows that the $2 \mu\text{m}$ square and disc as well as the smaller 500 and 400 nm squares have switched their central out-of-plane magnetization, e.g., from red to blue or vice versa. The size of the out-of-plane domain in the 400 nm

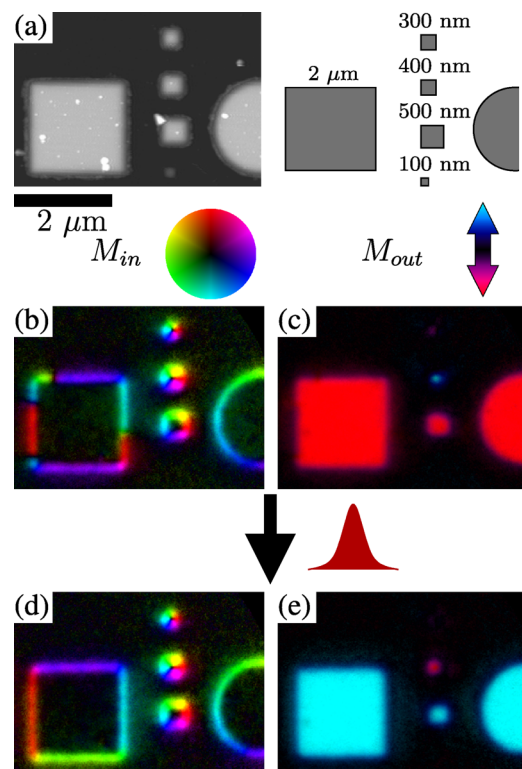


FIG. 2. (a) Atomic force microscopy image of the sample showing the $2 \mu\text{m}$ square and disc and the 500, 400, 300, and 100 nm squares. In-plane M_{in} and out-of-plane M_{out} magnetization images of the sample in its initial state (b) and (c) and after applying a single femtosecond laser pulse resulting in a slight change in the in-plane magnetization (d) while the out-of-plane magnetization reverses its direction, e.g., from down (red) to up (blue) magnetization direction (e).

square is found to be 200 nm. For the smaller 300 and 100 nm wide structures, their out-of-plane central domain could not be reliably imaged. The limit here arises from a combination of the instrument spatial resolution, the electric field distortion from the structures edges, and errors in the overlay of the three images recorded in different conditions.¹² At the same time, this azimuthal study reveals that all of the structures show an in-plane magnetized region of constant width at their edges. These in-plane magnetized rims explain the apparent multi-domain nature of the smaller structures observed in Figs. 1(b)–1(e), where the contrast is dominated by the in-plane magnetized domains. It is important to note that these in-plane rims do not play a role in the switching behaviour of the structures. This can be seen by comparing Figs. 2(b)–2(e) where, while the out-of-plane domains of the 500 and 400 nm structures switch, their in-plane rims remain the same. This illustrates experimentally the impetus by which this laser induced switching occurs. Indeed, simulations have shown that magnetic fields as high as 40 T were not sufficient to prevent the formation of the transient ferromagnetic-like state which leads to the magnetization reversal.³ This provides an insensitivity of the switched domains to their surroundings, a property which might be exploited to reach a very high recording density. There are cases where the in-plane magnetization changes after the laser excitations but does not influence the switching of the out-of-plane core, as can be seen for example at the edges of the 2 μm square in Fig. 2. Although the switching of the out-of-plane core is reproducible and the switching of in-plane structures has been demonstrated,³ the switching of the in-plane outer rim appears to be more complex. This might be related to the thinning, oxidation, and modified anisotropy at the edge of the structures. Furthermore, the laser intensity distribution is not homogeneous inside the structure due to laser interference within it.¹³ This difference in behavior of the edge and the core of the structures explains the apparently random switching of the small structures in Figs. 1(b)–1(e). In these images, the contrast of the small structures is dominated by the in-plane magnetization of the outer rim, and our azimuthal dependent study has allowed us to disentangle the switching behavior of the core and the outer rim.

Despite the insensitivity of the switching process of the magnetic domains to the surrounding magnetic configuration, it is nevertheless desirable to obtain mono-domain nanostructures for real device applications. It is thus important to understand the origin of the formation of the in-plane magnetized rims, which could be either an effect of oxidation from the uncapped structure edges or a structural change. To investigate possible oxidation of the structures, spectromicroscopy was performed at the Fe $L_{3,2}$ edges, for the center part and the outer edge of the structures. The resulting x-ray absorption spectrum (XAS) for a 400 nm wide disc is shown in Fig. 3(a). While the central region does not show oxidation, the edge region clearly contains a certain level of Fe^{3+} oxidation¹⁴ as shown by the additional shoulder visible in the spectrum and indicated by an arrow in Fig. 3(a). By computing the pixel-by-pixel difference between the Fe^{3+} and the Fe metal peaks images, a map of the oxidation is obtained and shown in Fig. 3(b). This reveals

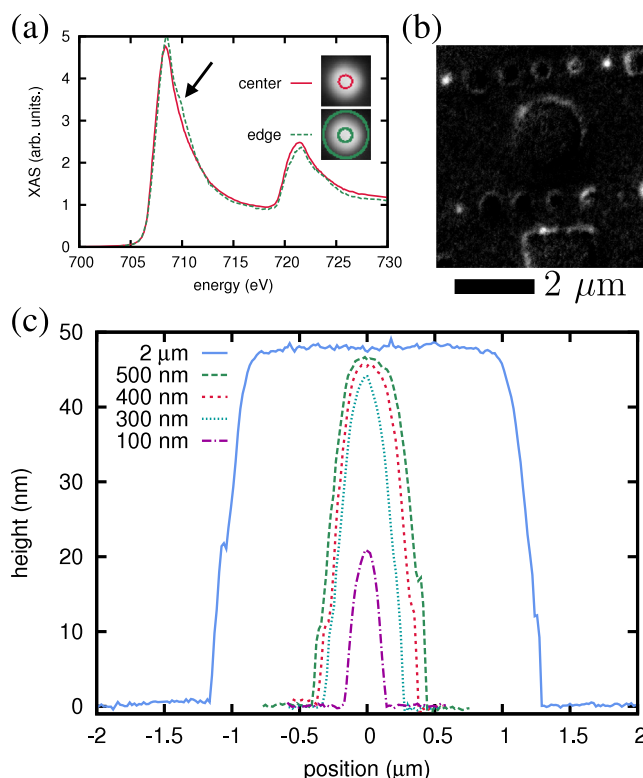


FIG. 3. (a) XAS spectrum obtained from the central and edge regions of a 400 nm wide disc, with the inset indicating the regions of interest. The arrow indicates the position of the Fe^{3+} oxide peak. (b) Oxidation image obtained by computing the pixel-by-pixel difference between the Fe oxide and metal peaks images. (c) AFM line profiles for the different structures.

that the oxidation is significantly inhomogeneous, with oxidation concentrated at certain positions around the structure edges, which appear as bright spots in Fig. 3(b). This observation is in conflict with the presence of the in-plane magnetized rims of constant width around the perimeter of the structures, suggesting that oxidation is not the main cause here. From the atomic force microscopy (AFM) image in Fig. 2(a), line profiles for each of the structures are extracted and are shown in Fig. 3(c). These line profiles reveal that a thinning of all the structures occurs at their edges over a distance of 150 nm. The lift-off process, which involves deposition of the magnetic layers on a structured polymer resist,⁷ results in thinner edges due to shadowing from the resist during film deposition. As the size of the structure becomes smaller than the shadow, this effect becomes more important to the point that it reduces the thickness of the 100 nm structures by a factor of 2. A reduced thickness of the GdFeCo film at the edges changes the magnetic anisotropy of the material. Since this edge thinning is occurring for all the structures, it correlates better with the in-plane magnetized rims than the inhomogeneous oxidation from the edges. In addition, this thinning of the edge over 150 nm would explain why the 300 nm wide and smaller structures have no central out-of-plane magnetized region, as shown in Fig. 2. This means that in lieu of the lift-off process used in this work, a subtractive etch process such as ion milling through a hard mask producing top-hat thickness profile should result in mono-domain nanostructures better suited for high density recording applications. Moreover, this edge thinning implies that the size of the central region of the 400 nm wide square

is consistent with the 200 nm domain size derived from the M_{out} component in Fig. 2(c). Such a domain size is particularly small considering that the average domain size in an unstructured GdFeCo film can be several tens of micrometer. This demonstrates the potential of the *heat pulse only* switching in structured recording media for future applications.

In conclusion, we have demonstrated laser induced magnetization switching in a 200 nm out-of-plane domain located in the center of a 400 nm wide nanostructure. The in-plane edge domains are found to play no particular role in the efficiency of the switching, thanks to the impetus by which the switching occurs and are likely the result of a thinning of the magnetic film due to the lift-off structuring method employed. This specific limitation can be lifted with an improved nanofabrication and optimized material properties leading to single domain structures with out-of-plane magnetization at sizes much smaller than the one reported here. This work constitutes an important step towards the combination of laser induced magnetization switching with bit patterned media for real device applications.

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